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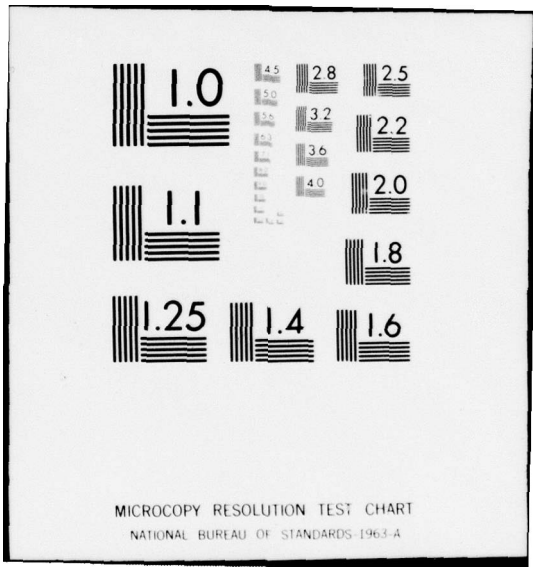
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Research and Development Technical Report

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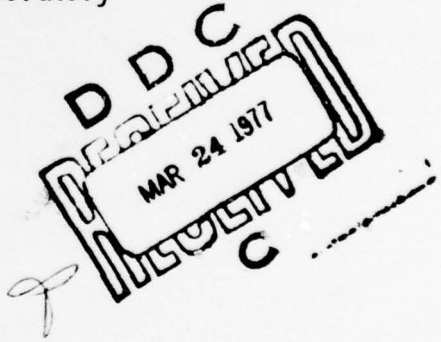
POLYCHROMATIC, FLASHLAMP-PUMPED DYE LASERS

T. F. Ewanizky

Combat Surveillance & Target Acquisition Laboratory



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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Techniques were investigated in an effort to accomplish two separate objectives with an unstable resonator flashlamp-pumped dye laser. First, the use of dye mixtures to determine whether laser action, at various visible wavelengths, could be simultaneously achieved from the different molecular species. Secondly, an optical system appropriate for broadband wavelength tunability of the unstable resonator laser was devised.

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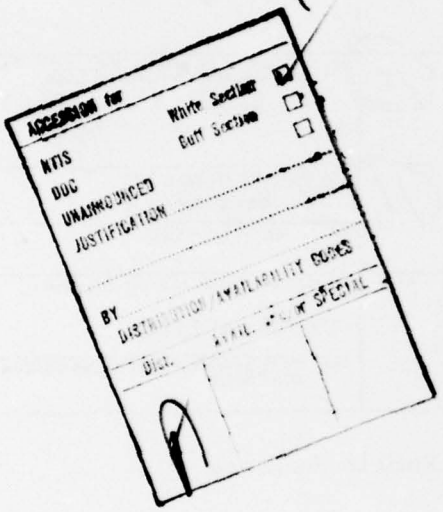
Results showed that polychromatic laser radiation from dye mixtures has limited feasibility at least with presently available dyes. However, simultaneous blue-green and orange laser radiation was obtained with a coumarin-rhodamine mixture. A continuous timing range of over 30 nm was efficiently achieved using a single-stage, rotatory dispersive intracavity filter with rhodamine 6G.



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POLYCHROMATIC, FLASHLAMP-PUMPED DYE LASERS

1. INTRODUCTION AND BACKGROUND

The purpose of this program was to examine methods to increase wavelength diversity in flashlamp-pumped organic dye lasers. Military relevance is connected to the Army need for wavelength-diversity in certain classified optical-countermeasure applications.

Two techniques were studied: First, laser solutions containing selected dye combinations were tested in an effort to find a combination which could provide polychromatic laser radiation. This had recently been demonstrated in a laser pumped system,[1] where as many as three different laser wavelengths in the blue, yellow and red were simultaneously emitted. The rationale of this program was to achieve similar results in a flashlamp-pumped system.

Secondly, a novel method for wavelength tuning the unstable resonator system[2] was devised. This resonator system was proven to be greatly superior to the previously utilized, conventional parallel-plane mirror cavities in its capacity for producing efficient dye laser operation with near diffraction-limited beam quality. Since the unstable resonator optics consist of spherical mirror surfaces, no simple arrangement of plane diffraction gratings or dispersing prisms is appropriate. The problem, therefore, was to design an optical tuning sub-assembly which could be simply inserted into the unstable resonator and effectively wavelength-tune the laser emission without disturbing the mode formation, or dynamic operation of the laser. That is, the tuning systems must essentially represent a variable wavelength transmitting filter which does not distort or deviate a beam propagating through it. Furthermore, it must have high transmission at the selected wavelength in order to ensure reasonable operating efficiency. In an effort to meet these requirements, the properties of a tuning system, whose principle is based on the rotatory dispersive property of crystalline quartz, was investigated with an unstable-resonator rhodamine 6G dye laser.

2. DYE MIXTURES

a. Experimental Results

Figure 1 shows the optical arrangement of the unstable-resonator dye laser system used in the experiments. The coaxial flashlamp and driver are commercial[3] with a maximum single-shot

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1. R. K. Jain and A. Dienes, Spectroscopy Letters 7, 491 (1974).
2. T. F. Ewanizky, Applied Physics Letters, 25, 295 (1974).
3. Phase-R Corporation, New Durham, NH

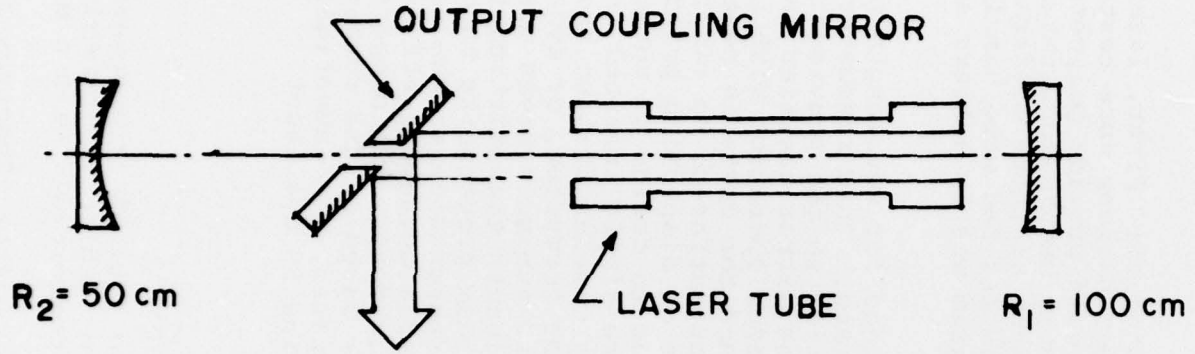

Figure 1. Optical configuration of the (negative branch) unstable resonator dye laser.

capability of 100 J input energy. The excited dye volume is 13 cm long by 1 cm diameter. Using an optical magnification of m = 2 gives a (geometric) output coupling of 75 percent from the angled mirror. The solution was circulated, from a polyethylene reservoir, by a variable speed centrifugal pump at a rate of 1 liter per minute. A millipore teflon filter was placed in series between the pump and flashlamp in order to reduce scattering centers due to bubbles or impurities.

Initial spectroscopic experiments consisted of making absorption spectra of various dyes on a Cary M18 absorption spectrophotometer, and recording fluorescence spectra with a Jarrel-Ashe f:6.3, plane-grating spectrograph. Compatible mixtures such as 7 diethylamino - 4 methyl coumarin (7d - 4m coumarin), which emits broadly in the blue, together with rhodamine 6G, which has a broad emission through the yellow to yellow-green, produced fluorescence which appeared virtually white.

Figure 2 shows the characteristic absorption spectra of these two dyes and the position of the laser emission from the combination. This typical figure illustrates the principle which allows simultaneous laser emission from the different species in a mixed dye solution. A first obvious requirement is that each dye must be soluble in a common solvent. In these experiments, only ethanol was used, not only because of good solubility but also because of inherent resistance to molecular aggregation and its non-toxicity. Secondly, optical interaction of the combination must be a minimum - that is, there can be no significant spectral overlap of the absorption spectrum of one specie with the fluorescence of another. Because of the broad electronic transitions typical of the organic dye molecules, this condition is generally not strictly possible.

Laser performance measurements were first made separately, and then in specific combinations of the most efficient laser dyes which span the visible spectrum. Input energies ranged to as high as 94 J, in an attempt to reach simultaneous laser threshold with the dye mixture solutions. The only successful dye combination proved to be that of 7d-4m coumarin and rhodamine 6G. The laser spectrum of this combination is shown in Figure 3.

It is interesting to note the double-band emission of 7d-4m coumarin. This phenomenon has been previously observed in other coumarin dyes and is ascribed to the transient formation of separate "exciplex" species from the neutral coumarin molecules.[4]

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4. A. Dienes, R. K. Jain and C. Lin, Applied Physics Letters, 22, 632 (1973).

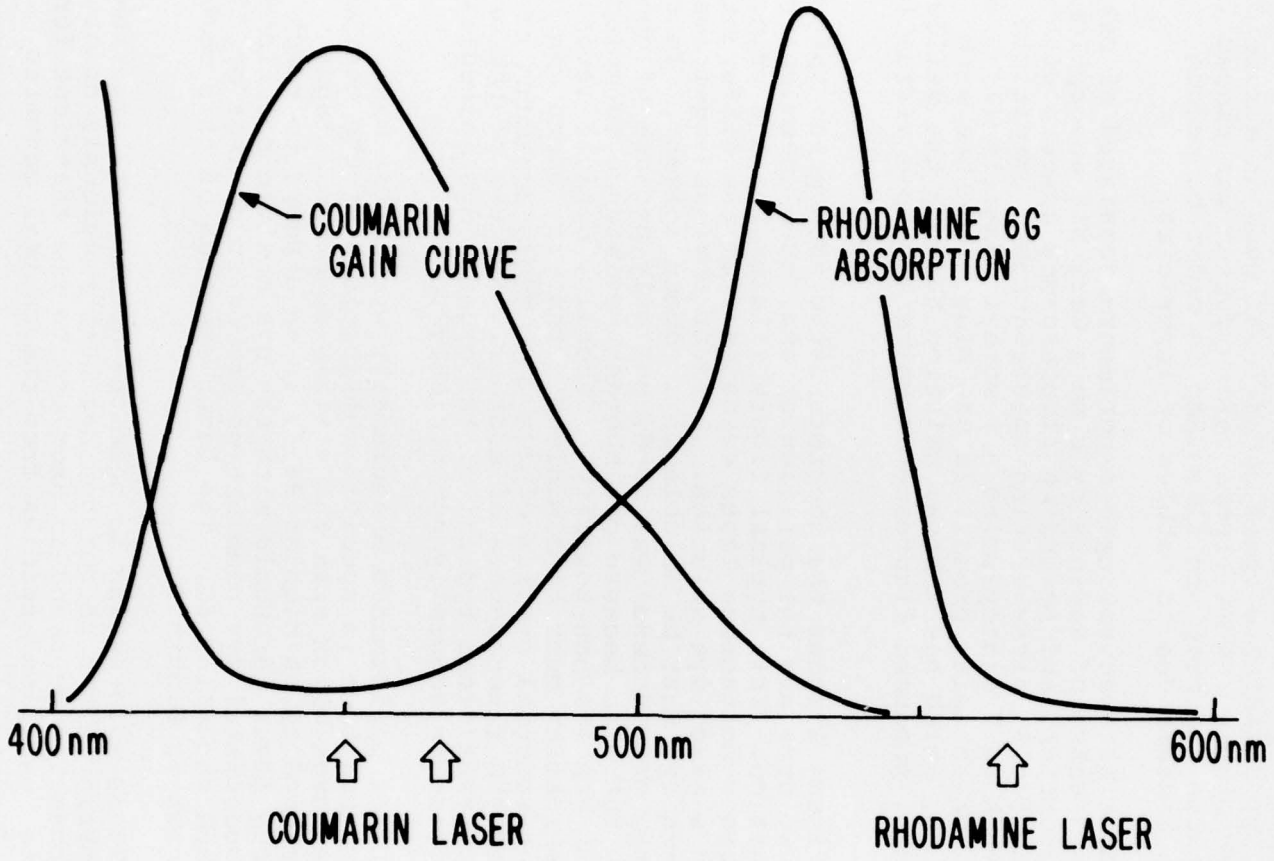

Figure 2. Characteristic absorption spectra of 7d-4m coumarin and rhodamine 6G, indicating laser wavelengths.

Figure 3. Simultaneous, blue and yellow-green laser emission from 7d-4m coumarin and rhodamine 6G. Mercury-cadmium calibration spectra are overlaid.

The general process that was followed in determining dye mixture concentration was as follows. First, a lasing concentration solution of the coumarin alone was prepared and its performance measured. Then, increasing concentrations of a secondary, yellow-green or red emitting dye was introduced and its effect on the laser performance of the primary dye was noted. Invariably, even with the addition of secondary dye concentrations too weak for secondary laser emission to occur, a substantial decrease in primary laser emission was observed. The problem thus resolved itself into arriving at a secondary dye concentration that was strong enough to lase, without substantially quenching laser emission from the primary dye due to absorption effects. Figure 4 shows the relative efficiencies of the coumarin and rhodamine 6G laser emission as a function of rhodamine concentration in the dye mixture. Note, that the best combined efficiency of this mixture occurs where the rhodamine concentration is about 2 orders of magnitude lower than that of coumarin. The blue and yellow-green simultaneous emission gave the visual appearance of white. (Experiments were carried out with opaque screens confining the beam, since no available sets of laser goggles for eye protection were effective against this spectral combination.)

Both rhodamine B and cresyl violet were tried in combination with 7d-4m coumarin, but no simultaneous laser action could be attained even with input energies approaching 100 J. In each case, coumarin emission was quenched before a lasing concentration of the secondary dye was attained. A tertiary addition of rhodamine 6G to the coumarin, cresyl-violet combination was tried in an effort to increase cresyl-violet gain via energy transfer from the rhodamine,[1] but these attempts were likewise unsuccessful.

To prove that these effects were due to only absorption by the secondary dye and not some chemical incompatibility, performance of a 7d-4m coumarin laser was measured with separate intra-cavity absorption cells containing various concentrations of rhodamine 6G solution. The same result, as occurred in the mixed solution, was noted.

b. Analysis and Discussion

A rate-equation analysis can be forwarded to explain the effects of absorption by the secondary dye. Although the effects of strong absorption on degrading laser performance is of course obvious, it appeared to be contradictory to intuition that relatively small absorption losses arising from the slight overlap of secondary dye concentrations could lead to such drastic consequences.

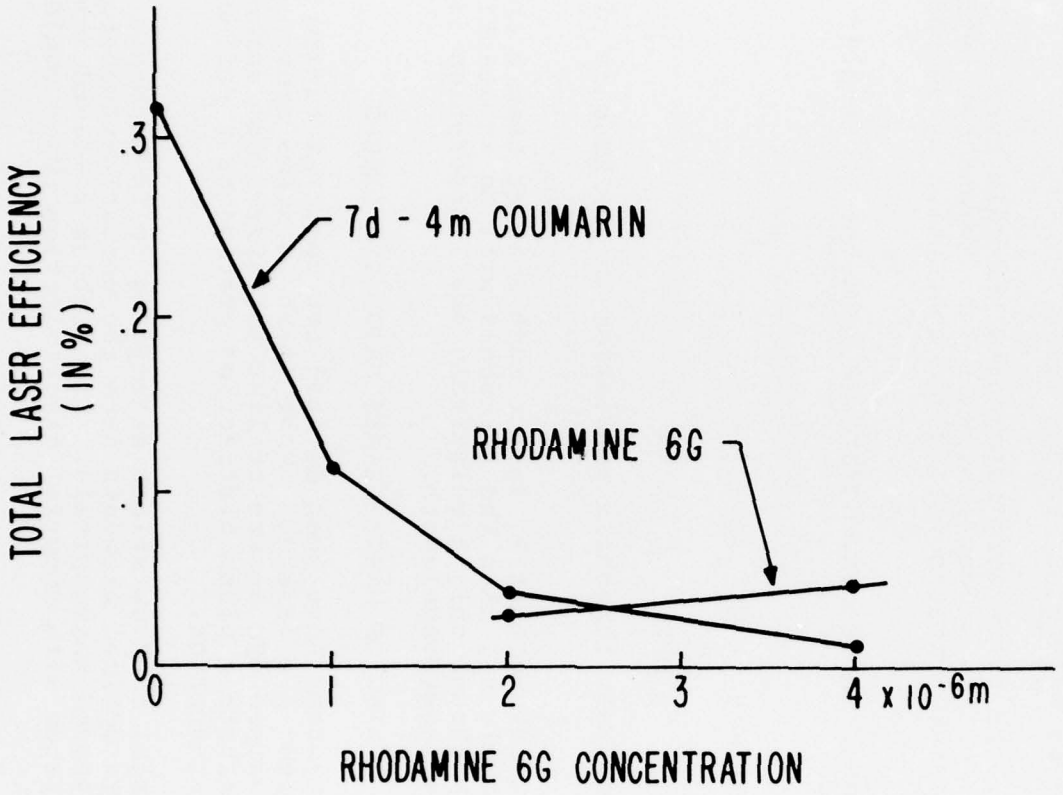

Figure 4. Total efficiencies of combination laser as a function of rhodamine 6G concentration. Values given are for 48.6 J input energies, and constant 7d-4m coumarin concentration of 2×10^{-4} m.

The rate of change of photons in the laser can be expressed as

$$dq(t)/dt = B \cdot n(t) \cdot q(t) - q(t)/t_c - B_A \cdot n_A \cdot q(t)$$

where $n(t)$ is the inverted population of the primary dye, n_A is the absorber population, t_c is the cavity lifetime, B and B_A are the Einstein coefficients for absorption by the laser and absorber molecules, respectively.

Thus, the first term represents the gain term responsible for photon production, while the second two represent losses due to laser output coupling and absorption by seconding dye molecules.

At laser threshold, $dq/dt = 0$, and the threshold population is

$$n_o' = 1/B \cdot t_c + B_A n_A/B$$

Or, $$n_o' = n_o + B_A n_{A/B},$$

where n_o is the laser threshold population in the absence of absorbers.

If we assume that B_A is not much smaller than B and n_A is approximately n_o, then the threshold will be substantially increased, even though only a relatively small absorption is observed at the laser wavelength.

3. SPECTRAL TUNING OF UNSTABLE-RESONATOR DYE LASERS

Numerous systems have been devised for spectral tuning of both CW and pulsed dye lasers.[5] Most of the systems appropriate for flashlamp-pumped dye lasers utilize multiple prism arrangements, or substitute a plane diffraction grating as a reflective element of the resonator.

Various trial configurations attempting to incorporate these dispersive optical elements into the unstable-resonator configuration proved unacceptable. The problem resolved itself to finding a system with spectral discrimination which could

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5. F. P. Schafer, Editor, "Dye Lasers," (Topics in Applied Physics, Vol 1, Springer - Verlag Publishers, 1973).

be placed directly into the unstable-resonator without optically perturbing the resonator mode. The tuning device reported by Kato and Sato[6] was investigated for this purpose. Figure 5 illustrates how the rotatory dispersive filter elements were incorporated into the unstable resonator.

The principle of the rotatory dispersive filter is based on the property of crystalline quartz (and some other materials) of producing a rotation of the plane of polarization of a light beam.[7] The degree of rotation is a function of wavelength, so that if a z-cut plate of crystalline quartz is placed between two polarizers, the combination will show a spectrally periodic transmission. The transmission wavelengths may be continuously varied by changing the relative orientation angle of the polarizers, and thus tuning the filter. Figure 6 shows a spectral recording of white light transmitted through a filter composed of two laser-grade polarizers and a 2.3 cm long plate of crystalline quartz cut with z-axis parallel to the axis of transmission. A channeled spectrum results with broad, periodic intensity maxima.

Figure 7 shows the tuning effect produced with the unstable resonator dye laser. A continuous tuning range of over 300 Å, from approximately 5685 Å in the yellow-green to 6020 Å in the orange, could be attained with a 120 degrees total rotation of the tuning polarizer, using a 1×10^{-5} m rhodamine 6G, ethanol solution. A similar tuning range was obtained with higher concentrations, but self-absorption effects tended to shift the spectra to slightly longer wavelengths.

It is interesting to note the effect of tuning on the laser bandwidth. The untuned laser emits at a center wavelength of approximately 5730 Å, with a bandwidth of approximately 80 Å. However, when tuned away from the gain peak to a center wavelength of about 5850 Å, the bandwidth increases to over 175 Å. Presumably, this is due to a combined effect of the broadness of both the filter and gain curve bandwidth.

Figure 8 shows the change in output energy of the laser when tuned through its spectral range. The output energy and spectral position of the untuned laser are plotted for comparison. Note that a remarkably small difference in laser efficiency occurs at the untuned wavelength, with the laser operating untuned or with addition of the tuning optics. In fact, no large fluctuations occur over the whole tuning range. A change of only about $\pm$ 20 percent from the main output energy occurs over the whole tuning range.

6. D. Kato and T. Sato, Optics Communications, Vol 5, p 134 (1972).

7. R. W. Ditchburn "Light," by R. W. Ditchburn (2nd Edition, Interscience Publishers, NY), p 486.

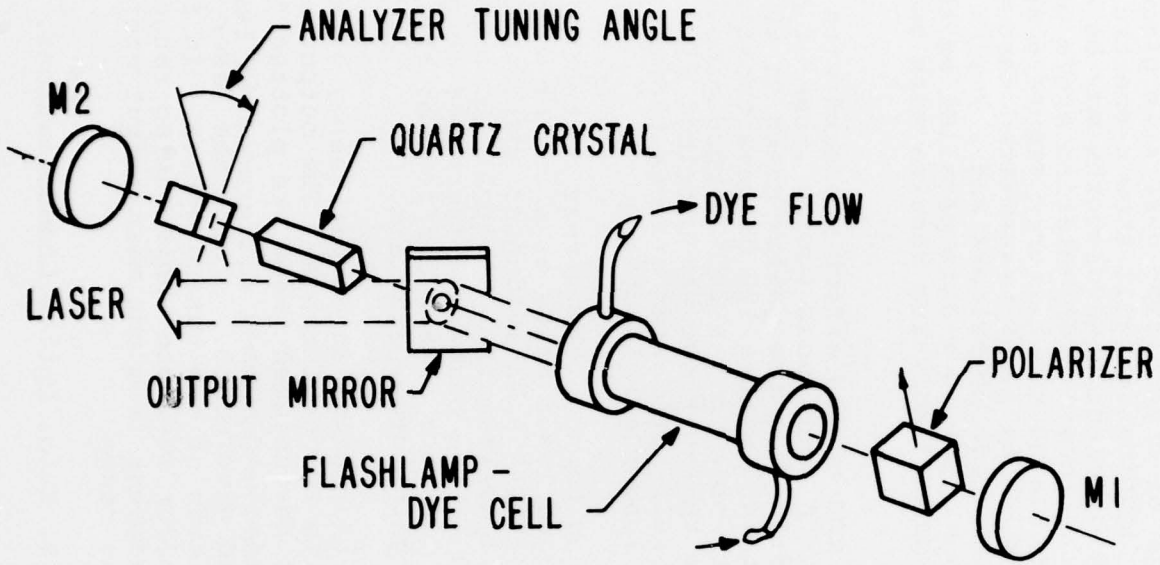

Figure 5. Optical arrangement of the unstable resonator with the rotatory dispersive filter.

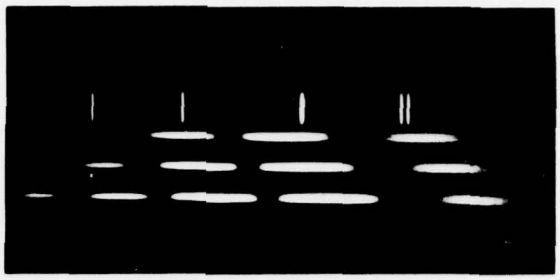

Figure 6. Channeled spectra produced by rotatory dispersive filter. Spectral location of transmission maxima and minima are shown to be varied in changing relative polarizer orientation in increments of 30°. A mercury-cadmium calibration spectrum is shown at top for reference.

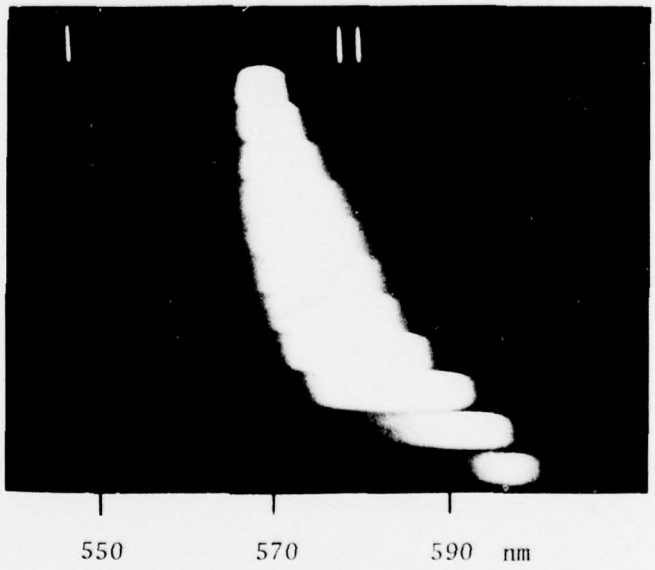

Figure 7. Spectrally tuned dye laser emission. Spectra, from top to bottom, each correspond to progressive 10° increments of increased analyzer orientation angle.

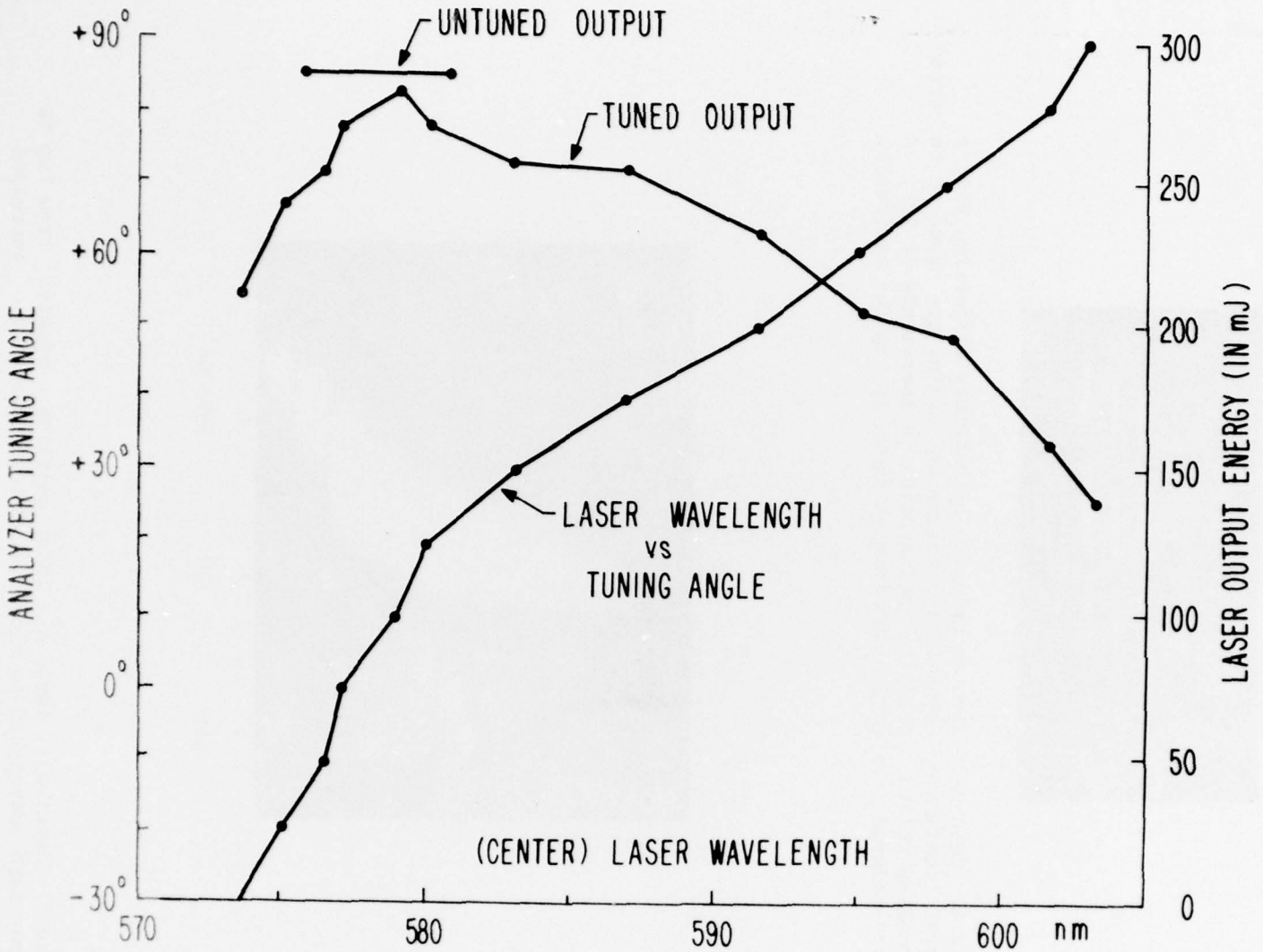

Figure 8. Output energy and central wavelength of the spectrally tuned rhodamine 6G dye laser, as a function of analyzer tuning angle. The results shown are for single-shot, 60 J (electrical) input energy pulses.

Another interesting effect occurs if a simple etalon is additionally introduced into the resonator. A thin, uncoated sheet of mica, peeled from a plate of unknown optical quality, was mounted near the short focal-length mirror and produced the results shown in Figure 9. Tuning range and output energy were essentially unaffected, but multiple rather than broadband emission occurred at wavelengths separated by the free spectral-range of the etalon.

4. RESULTS AND CONCLUSIONS

The use of organic dye mixtures to produce multiple wavelength laser emission appears to be limited in scope, at least with dyes that are commonly available.

Results of tuning an unstable-resonator dye laser with a single-stage rotatory dispersive filter were promising and could be relevant for military application. High operating efficiency and a broad spectral tuning range from the yellow-green to orange could be easily attained in spite of the use of non-optimizd, uncoated optics. The beneficial qualities of the unstable resonator configuration is amply demonstrated in this regard. In spite of the strong reflections inevitably due to the uncoated optical surfaces, the unstable resonator mode was clearly dominant since no near-field hot spots, indicating parasitic oscillation, were observed.

The most straightforward approach to producing narrower bandwidth, tunable radiation, would appear to be the adoption of a two-stage rotatory dispersive filter as described by D. Kato and T. Sato.[6] With proper, compact construction and anti-reflection coated optics, laser bandwidth should be substantially narrowed without undue sacrifice of operating efficiency.

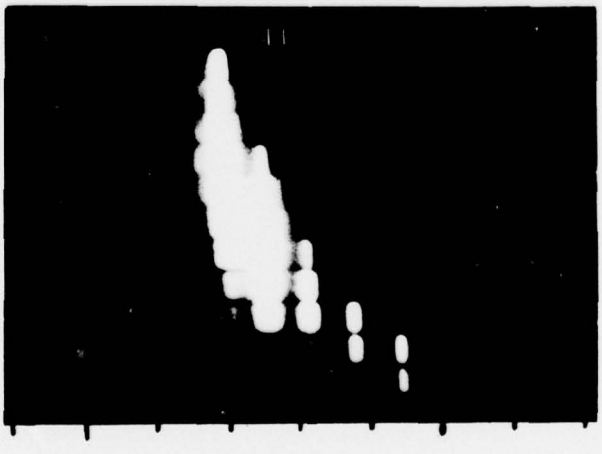

Figure 9. Spectrally tuned rhodamine 6G laser emission as in Figure 7, but with a thin, uncoated mica etalon additionally introduced into the resonator.